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Oblique-incidence optical reflectivity difference from a rough film of crystalline material

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Formation of a rough film of crystalline material on a smooth substrate resulting from kinetic roughening in epitaxy or erosion causes disproportionate changes in reflectivity for *s*- and *p*-polarized light. I present a mean-field theory of optical reflectivity difference defined as $(r_p - r_{p0})/r_{p0} - (r_s - r_{s0})/r_{s0} \equiv \Delta_p - \Delta_s$ from such a rough film, with r_{p0} and r_{s0} being the reflectivities of the bare substrate, and r_p and r_s being the reflectivities after the rough film forms on the substrate. In the limit that the average film thickness is less than the optical wavelength λ, I found that $\Delta_p - \Delta_s$ consists of a term that varies linearly with the average film thickness and a term that is proportional to the surface density of step edge atoms. I apply such a theory to the analysis of growth and ion erosion of a number of crystalline materials studied with the oblique-incidence optical reflectivity difference (OI-RD) technique.

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Epitaxial growth and removal of crystalline materials under various physical/chemical conditions are among the main topics of material sciences. Experimental capabilities of monitoring the morphology of a growth or erosion surface are crucial to characterization, understanding, and in turn control of growth and erosion processes.[1–8] Recently an oblique-incidence optical reflectivity difference technique, a special form of nulling ellipsometry, has been applied to studies of a wide range of surface kinetic processes in vacuum and in electrochemical environment.[9–13] Unlike electron diffraction techniques such as RHEED or helium scattering techniques,[1–7] the optical technique is capable of probing growth or erosion surfaces under high ambient pressure or in liquids as well as under ultrahigh vacuum. In addition the optical response from a surface is sensitive to both crystalline order and chemical make-up of the surface. Such a dual sensitivity has been exploited in experimental studies of both growth and surface reaction kinetics in rare gas and perovskite oxide epitaxy.[9–12]

So far the optical response is treated with a three-layer model in a mean-field sense.[14] Such a model is sensible if each molecular layer of the growth surface can be treated as a uniform mixture of host and guest materials with their respective bulk optical dielectric constants. When the surface roughness of an otherwise crystalline material extends beyond one monolayer, the three-layer model or the modification of the model by Aspnes and co-workers[15] assumes that the atoms including voids within each atomic/molecular layer at same height from the substrate experiences same mean electric field, and the dielectric response of each layer is a volume average of those of bulk atoms and voids within the layer based on a self-consistent effective medium approximation (EMA) proposed by Bruggeman.[16] The contributions from surface atoms including those at step edges are either neglected or not treated explicitly. Though convenient and in some cases successful,[15,17] such an assumption is questionable voids are not inclusions and thus the electric field in a void is expected to be different from that in the corresponding filled region. In addition atoms or molecules at interfaces and particularly at step edges have distinctly different dielectric responses. When their contributions are no longer negligible, the modified three-layer model with Bruggeman's effective medium approximation does not lend itself to a transparent analysis of such contributions. In early stages of growth or erosion, the variation of step edge density is one of the most useful indicators of whether the processes proceed in a layer-by-layer fashion (i.e., two-dimensionally), or a three-dimensional island fashion, or otherwise.[10,18]

In this paper, I propose a different mean-field model for dealing with optical reflection from a rough film of crystalline material with characteristic roughness length scales much less than optical wavelengths. In this model, I compute the sum of reflection from flat terraces of the surface, each of which consists of terrace atoms and step edge atoms (with their respective dielectric constants), a layer of bulk atoms that may be different from the substrate, and the underlying substrate. This model treats voids naturally under the circumstances. The main result of this model is that *the optical reflectivity difference has a term that is proportional to the mean thickness of the rough film and a term that varies linearly with the density of step edge atoms*. This model makes the correlation of the optical reflectivity difference, measurable during thin film growth and erosion, to the surface morphology more straightforward and transparent.

As shown for example by Kalff *et al.*,[19] Vrijmoeth *et al.*,[20] and others in scanning tunneling microscopy studies of epitaxy and ion erosion of crystalline metals, a majority of surface atoms on a growth or erosion surface are on terraces and a minority of them are at step edges. In this case the net reflection of a light can be considered as the sum of the reflection from all terraces at different heights from the substrate surface plane as I will justify shortly. Since the dimension of each terrace segment on a rough film is much smaller than optical wavelengths, the radiation from such a segment *alone* would be diffusive. However due to the fact that the characteristic dimension and separation between rough features are small compared to optical wavelengths, only the

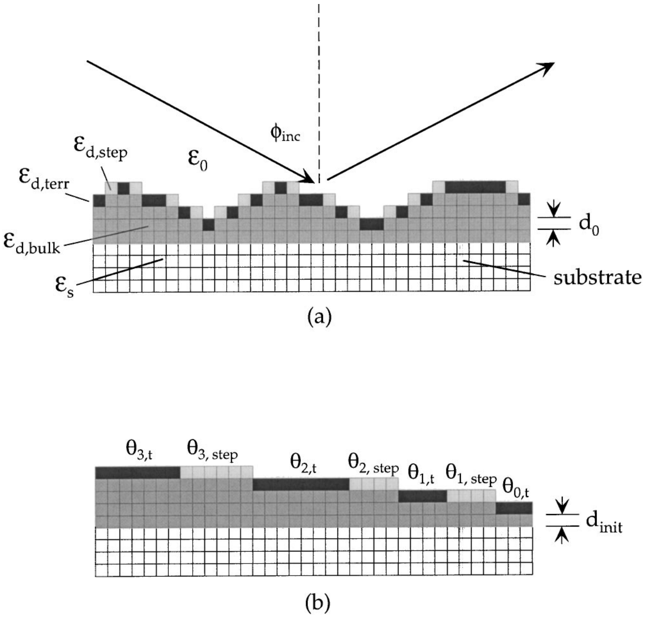

FIG. 1. (a) Sketch of a rough crystalline thin film on top of a smooth substrate (white squares, characterized by ε_s). Dark gray squares: atoms inside the rough film, characterized by $\varepsilon_{d,\mathrm{bulk}}$. Black squares: terrace atoms, characterized by $\varepsilon_{d,\mathrm{terr}}$. Light gray squares: step edge atoms, characterized by an effective $\varepsilon_{d,\mathrm{step}}$. (b) The rearrangement of the rough crystalline film for the purpose of computing specularly reflected light.

radiation in specular direction survives the summation over the contributions from all terrace segments. Therefore we should only concern ourselves with the radiation from each terrace segment in specular direction and in the associated direction inside the rough surface.

The concept of considering the reflection from a terrace segment locally is justified if the reflected radiation from the segment results only from the local structure and dielectric properties. Consider reflection from a three-layer system as in a classical Fabry–Perot interferometer.[21] The reflection is governed by the dielectric property over a range determined by the effective number (N) of multiple reflections in a film of thickness h. N is given by $N=4\sqrt{R_{21}R_{23}}/(1-\sqrt{R_{21}R_{23}})$, where R_{21} and R_{23} are reflectances (absolute squares of reflectivity) of light from the interfaces between the film (medium #2) and the ambient (medium #1) and between the film and the substrate (medium #3), respectively. At optical wavelengths, $\sqrt{R_{21}R_{23}}\sim 0.5$, one has $N\sim 4$. Since the lateral distance traveled by the light between two successive reflections is roughly $\delta x=(\sin\phi_{\mathrm{inc}}/\sqrt{|\varepsilon_d|})(h/2)\sim h/12$, the linear size of the region that contributes to a local reflection is roughly $N\delta x\sim h/3$. For a rough film of thickness $h=10$ layers, the reflection from a terrace segment can be considered *local* if the width of the terrace is more than that of 3 layers. This holds true during initial stages of a film growth or erosion. The morphology of a rough crystalline film at these early stages already enables determination of the kinetics of the process. It is this limit that the present model will be most useful.

In Fig. 1(a), I show the sketch of a rough film on a smooth substrate (ε_s). The film is in contact with a lossless ambient with an optical dielectric constant ε_0. The atoms inside the film are characterized by a bulk-phase optical dielectric constant $\varepsilon_{d,\mathrm{bulk}}$. The atoms inside terraces of the outermost layer are characterized by a different dielectric constant $\varepsilon_{d,\mathrm{terr}}$. The atoms at step edges are characterized by an effective dielectric constant $\varepsilon_{d,\mathrm{step}}$. I should note that the reflection from each terrace segment contains separate contributions from the terrace atoms and the step edge atoms. Not only the dielectric response from the latter is different from that of the terrace atoms, the effective electric field experienced by the step edge atoms is also different and somewhat ill-defined. I will treat this electric field in a mean-field sense and incorporate the unaccounted effect into $\varepsilon_{d,\mathrm{step}}$.

Let a collimated beam of light with wavelength λ incident on the rough film as shown in Fig. 1(a) at angle ϕ_{inc}. Since the incident light needs to reach different depths for terraces at different heights from the substrate, and the reflected radiation in specular direction from different terraces travel different distances before they add to yield the net reflection, it is necessary to keep track of the height of a terrace segment under consideration. For terrace segments at the same height from the substrate surface, the sum of the distance covered by the incident light from a reference wave front and the distance covered by the specular reflected light to another reference wave front on the reflection side is a constant. It is thus sensible to treat the radiation from those terrace segments at a fixed height from the substrate surface together. For this purpose I rearrange the rough film by grouping together the terraces at same height and furthermore separating each of the "coalesced" terraces into a region of terrace atoms and a region of step edge atoms, as shown in Fig. 1(b).

Let θ_j be the coverage of the jth "coalesced" terrace at a distance $d_j=jd_0+d_{\mathrm{init}}$ away from the substrate surface. d_0 is the thickness of one monolayer in the direction normal to the surface, and d_{init} is the smallest height of the rough film from the substrate surface. Let $\theta_{j,t}$ be the coverage of terrace atoms, $\theta_{j,\mathrm{step}}$ be the coverage of step edge atoms, and $\theta_j=\theta_{j,t}+\theta_{j,\mathrm{step}}$. It is clear that $\Sigma_{j=0}\theta_j=\Sigma_{j=0}\theta_{j,t}+\Sigma_{j=0}\theta_{j,\mathrm{step}}=1$. I define the total coverage of terrace atoms as $\theta_t\equiv\Sigma_{j=0}\theta_{j,t}$, and the total coverage of step edge atoms as $\theta_{\mathrm{step}}\equiv\Sigma_{j=0}\theta_{j,\mathrm{step}}$.

The reflectivity for s- and p-polarized light from such a rough film can be written as the sum of contributions from all terraces,

$$r_{s(p)}=\sum_{j=0} r^{(t)}_{s(p)}(d_j)\,\theta_{j,t}\exp(-i4\pi d_j\cos\phi_{\mathrm{inc}}/\lambda)$$

$$+\sum_{j=0} r^{(\mathrm{step})}_{s(p)}(d_j)\,\theta_{j,\mathrm{step}}\exp(-i4\pi d_j\cos\phi_{\mathrm{inc}}/\lambda), \qquad (1)$$

$r^{(t)}_{s(p)}(d_j)$ is the reflectivity for $s(p)$-polarized light from a three-layer system that consists of a monolayer of terrace atoms, a layer of bulk-phase atoms with a thickness of d_j-d_0, and the substrate. $r^{(\mathrm{step})}_{s(p)}(d_j)$ is the reflectivity from a similar three-layer system in which the topmost monolayer is replaced by atoms characterized by $\varepsilon_{d,\mathrm{step}}$. In the limit that the thickness of the rough film is much less than the wavelength, the correction to the reflectivity from a bare substrate is small and varies linearly with the thickness of the rough film. In this case the contributions from the topmost layer and the bulk-phase layer are additive, and $r^{(t)}_{s(p)}(d_j)$ and $r^{(\mathrm{step})}_{s(p)}(d_j)$ are given by[9–13]

$$r^{(t)}_{s(p)}(d_j)=r_{s(p)0}[1+\Delta^{(\mathrm{bulk})}_{s(p)}(d_j-d_0)+\Delta^{(t)}_{s(p)}(d_0)], \quad (2)$$

$$r^{(\mathrm{step})}_{s(p)}(d_j)=r_{s(p)0}[1+\Delta^{(\mathrm{bulk})}_{s(p)}(d_j-d_0)+\Delta^{(\mathrm{step})}_{s(p)}(d_0)]. \quad (3)$$

Inserting Eqs. (2) and (3) into Eq. (1) and keeping only terms that vary linearly with the thicknesses,

$$\begin{aligned} r_{s(p)}=r_{s(p)0}\sum_{j=0}&[1+\Delta^{(\mathrm{bulk})}_{s(p)}(d_j-d_0)+\Delta^{(t)}_{s(p)}(d_0)\\ &-i4\pi d_j\cos\phi_{\mathrm{inc}}/\lambda]\theta_j+r_{s(p)0}\\ &\times[\Delta^{(\mathrm{step})}_{s(p)}(d_0)-\Delta^{(t)}_{s}(d_0)]\theta_{\mathrm{step}}, \end{aligned} \quad (4)$$

I define the optical reflectivity difference as

$$\Delta_p-\Delta_s\equiv\frac{r_p-r_{p0}}{r_{p0}}-\frac{r_s-r_{s0}}{r_{s0}}. \quad (5)$$

From Eq. (4), I arrive at

$$\begin{aligned} \Delta_p-\Delta_s=\sum_{j=0}&[\Delta^{(\mathrm{bulk})}_{p}(d_j-d_0)-\Delta^{(\mathrm{bulk})}_{s}(d_j-d_0)]\theta_j\\ &+[\Delta^{(t)}_{p}(d_0)-\Delta^{(t)}_{s}(d_0)]\theta_t\\ &+[\Delta^{(\mathrm{step})}_{p}(d_0)-\Delta^{(\mathrm{step})}_{s}(d_0)]\theta_{\mathrm{step}}. \end{aligned} \quad (6)$$

Zhu and co-workers have shown that[9–12]

$$\Delta^{(t)}_{p}(d_0)-\Delta^{(t)}_{s}(d_0)=\alpha d_0\left[\frac{(\varepsilon_{d,\mathrm{terr}}-\varepsilon_0)(\varepsilon_{d,\mathrm{terr}}-\varepsilon_s)}{\varepsilon_{d,\mathrm{terr}}}\right], \quad (7)$$

$$\Delta^{(\mathrm{step})}_{p}(d_0)-\Delta^{(\mathrm{step})}_{s}(d_0)=\alpha d_0\left[\frac{(\varepsilon_{d,\mathrm{step}}-\varepsilon_0)(\varepsilon_{d,\mathrm{step}}-\varepsilon_s)}{\varepsilon_{d,\mathrm{step}}}\right], \quad (8)$$

$$\begin{aligned} &\Delta^{(\mathrm{bulk})}_{p}(d_j-d_0)-\Delta^{(\mathrm{bulk})}_{s}(d_j-d_0)\\ &\quad=\alpha\times(d_j-d_0)\left[\frac{(\varepsilon_{d,\mathrm{bulk}}-\varepsilon_0)(\varepsilon_{d,\mathrm{bulk}}-\varepsilon_s)}{\varepsilon_{d,\mathrm{bulk}}}\right], \end{aligned} \quad (9)$$

$$\alpha\equiv(-i)\frac{4\pi\cos\phi_{\mathrm{inc}}\sin^2\phi_{\mathrm{inc}}\sqrt{\varepsilon_0}\varepsilon_s}{\lambda(\varepsilon_s-\varepsilon_0)(\varepsilon_s\cos^2\phi_{\mathrm{inc}}-\varepsilon_0\sin^2\phi_{\mathrm{inc}})}. \quad (10)$$

Since $\Sigma_{j=0}(d_j-d_0)\theta_j=\langle d\rangle-d_0$ with $\langle d\rangle=\Sigma_{j=0}d_j\theta_j$ being the average thickness,

$$\begin{aligned} \Delta_p-\Delta_s=\alpha&\left[\frac{(\varepsilon_{d,\mathrm{bulk}}-\varepsilon_0)(\varepsilon_{d,\mathrm{bulk}}-\varepsilon_s)(\langle d\rangle-d_0)}{\varepsilon_{d,\mathrm{bulk}}}\right.\\ &\left.+\frac{(\varepsilon_{d,\mathrm{terr}}-\varepsilon_0)(\varepsilon_{d,\mathrm{terr}}-\varepsilon_s)d_0}{\varepsilon_{d,\mathrm{terr}}}\right]\\ &+\alpha d_0\left[\frac{(\varepsilon_{d,\mathrm{step}}-\varepsilon_0)(\varepsilon_{d,\mathrm{step}}-\varepsilon_s)}{\varepsilon_{d,\mathrm{step}}}\right.\\ &\left.-\frac{(\varepsilon_{d,\mathrm{terr}}-\varepsilon_0)(\varepsilon_{d,\mathrm{terr}}-\varepsilon_s)}{\varepsilon_{d,\mathrm{terr}}}\right]\theta_{\mathrm{step}}. \end{aligned} \quad (11)$$

Equation (11) is the main result of this paper. In homoepitaxy or erosion processes, the bulk-phase film is the same as the substrate $\varepsilon_{d,\mathrm{bulk}}=\varepsilon_s$. As a result the first term in the first bracket of Eq. (11) drops off. The model predicts that the optical reflectivity difference is proportional to the density of step edge atoms θ_{step}. In a step-flow growth or erosion where θ_{step} remains unchanged, $\Delta_p-\Delta_s$ is expected to remain constant as well. In a layer-by-layer growth where θ_{step} oscillates with continuous deposition, $\Delta_p-\Delta_s$ is expected to oscillate accordingly just as RHEED or helium scattering.[1–7] I should note that in a reactive molecular beam epitaxy, atomic constituents are deposited sequentially such that the chemical make-up and the resultant $\varepsilon_{d,\mathrm{terr}}$ of the topmost molecular layer go through oscillatory changes as each layer of unit cells is formed. As a result, an additional oscillatory variation in $\Delta_p-\Delta_s$ originated from $\varepsilon_{d,\mathrm{terr}}$ is expected.[2,7,8] In a three-dimensional growth (island growth) or erosion when θ_{step} increases monotonically, $\Delta_p-\Delta_s$ is expected to vary monotonically accordingly. In heteroepitaxy, in addition to the dependence on θ_{step}, $\Delta_p-\Delta_s$ also increases (or decreases) linearly with the average thickness $\langle d\rangle$ of a rough film. These behaviors have been observed experimentally.

I now use the present model to examine the findings of a number of OI-RD experiments in which complex $\Delta_p-\Delta_s$ have been determined directly. The experimental setup for an oblique-incidence optical reflectivity difference technique has been described in details recently in Refs. 9–12. As a quantitative test for such a mean-field model, Landry and co-workers have measured $\Delta_p-\Delta_s$ from one and two monolayers of Xe films on Nb(110).[22] By comparing with the first term in Eq. (11) using $\varepsilon_s=-0.25+i16.13$, $\varepsilon_{d,\mathrm{terrace}}\approx\varepsilon_{d,\mathrm{bulk}}=2.19$, and $d_o=3.55$ Å, they found that the model reproduced the incidence-angle dependence and the magnitude of the experimental $\Delta_p-\Delta_s$.

OI-RD studies of kinetic roughening in rare-gas epitaxy. Recently Nabighian *et al.*[10] and Thomas *et al.*[12] reported studies of Xe growth on Ni(111) and Nb(110) using the techniques of OI-RD and LEED. On Ni(111) the first monolayer of Xe forms a commensurate, nearly strain-free $(\sqrt{3}\times\sqrt{3})R30°$ (hexagonal close-packed) structure. The subsequent Xe growth is a model case of vapor-phase homoepitaxy on the (111) plane of a fcc crystal. Nabighian and co-workers observed that at 40 K, the growth of Xe on the $(\sqrt{3}\times\sqrt{3})R30°$-Xe superlattice proceeds in a layer-by-layer mode and the corresponding $\mathrm{Im}\{\Delta_p-\Delta_s\}$ varies periodically with Xe exposure. It is noteworthy that the oscillations can only originate from a periodic change in θ_{step}. When the substrate temperature is reduced to 35 K, the Xe growth becomes rough (or three-dimensional), and the corresponding $\mathrm{Im}\{\Delta_p-\Delta_s\}$ was found to vary monotonically with Xe exposure. Both are consistent with the prediction of Eq. (11). On Nb(110), the first two Xe monolayers form a transition layer to accommodate the lattice mismatch and symmetry frustration between the (110) plane of a bcc metal and the (111) plane of a Xe crystal. The third monolayer of Xe forms a bulk-like hexagonal close-packed structure. Thomas and co-workers observed that between 53 and 47 K, the Xe growth on top of the third Xe monolayer proceeds in a step-flow mode and the corresponding $\Delta_p-\Delta_s$ change monotonically with Xe exposure. At around 40 K, the growth mode goes through a transition from a step-flow growth to a layer-by-layer growth, signified by a small oscillatory component on top of the monotonic envelope of $\mathrm{Re}\{\Delta_p-\Delta_s\}$. Although the signal-to-noise ratio was limited, the line shape of the oscillation roughly reproduced the functional form, $\sim(1-\theta)\sqrt{-\ln(1-\theta)}$, predicted by Stoyanov and Michailov for θ_{step} for a layer-by-layer growth (through homogeneous nucleation and growth of compact 2D islands).[23] Below 35 K, the growth becomes rough and the oscillatory component in $\mathrm{Re}\{\Delta_p-\Delta_s\}$ vanishes. More recently, Fei and co-workers also observed oscillations in $\Delta_p-\Delta_s$ in response to a layer-by-layer growth of Nb-doped $\mathrm{SrTiO_3}$ on $\mathrm{SrTiO_3}(001)$.[24] These authors found the oscillations to persist over hundreds of monolayers.

OI-RD studies of ion erosion of crystalline metals. Erosion of crystalline metals can be considered as an inverse process of homoepitaxy in which atoms are removed from a surface instead of being added to the surface. In an ion erosion process, vacancies (created by ion sputtering of surface atoms off flat terraces) play the similar role as deposited adatoms do in homoepitaxy. Depending upon the mobility of surface vacancies, the erosion of a crystalline surface may proceed in a step-flow mode, or a layer-by-layer mode, or a three-dimensional mode in which case multilayer mounds and pits form. As shown by Kalff *et al.*,[19] the slope of a multilayer mound or pit is a characteristic of the balance between the erosion and the kinetic-limited annealing process. Since the slope of a mound or pit is proportional to the density of step edge atoms, the OI-RD signal from an eroding surface *directly* measures the average slope. In Fig. 2, I display $\mathrm{Re}\{\Delta_p-\Delta_s\}$ from an Nb(110) surface that is eroded by 1-keV Ar/Ne ions. Two monolayers of Nb are removed after 600 s. At 1073 K, $\mathrm{Re}\{\Delta_p-\Delta_s\}$ remains unchanged, indicating that the erosion proceeds in a step-flow mode. As the temperature decreases, $\mathrm{Re}\{\Delta_p-\Delta_s\}$ increases monotonically with ion exposure, indicating that the roughness builds up, and thus θ_{step} increases. Since θ_{step} is proportional to the slope, $\mathrm{Re}\{\Delta_p-\Delta_s\}$ shows the evolution of the average slope. I note that although scanning tunneling microscopy reveals atomic scale details of a rough surface,[19,20] it is not a convenient experimental method for extracting ensemble averaged quantities of a rough surface since extensive data have to be taken and analyzed. In this regard, the technique of OI-RD has an inherent advantage of measuring directly and continuously the average slope on a rough surface with high accuracy. The latter is crucial for studying scaling behaviors of erosion or three-dimensional growth.

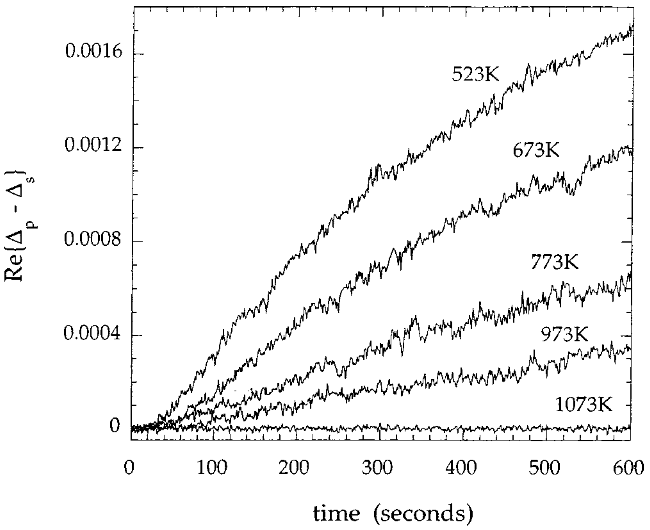

FIG. 2. Evolution of the real part of oblique-incidence optical reflectivity difference, $\mathrm{Re}\{\Delta_p-\Delta_s\}$, from an Ar/Ne ion-eroded Nb(110) surface at different temperatures. On average two monolayers of Nb atoms are removed after 600 s.

In conclusion, when the mean thickness of a rough crystalline film on a smooth substrate is small compared to optical wavelengths, and when the average slope of the rough features is small, the net optical reflection from such a surface is approximately the sum of reflections from terrace segments of the surface. As a result, the experimentally measurable reflectivity difference, defined as $\Delta_p-\Delta_s$, consists of a term that is proportional to the mean thickness of the bulk-phase portion of the film, a term that is a function of the dielectric response of terrace atoms (or unit cells), and a term that is proportional to the density of step edge atoms. In the limit that this model applies, it offers a transparent mean to analyze the morphology of a growth or eroding surface in real time and is thus effective for studies of growth and erosion kinetics under a wide range of conditions that are accessible by light.

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